

Quantum Limit in a Parallel Magnetic Field in Layered Conductors

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Abstract

We show that electron wave functions in a quasi-two-dimensional conductor in a parallel magnetic field are always localized on conducting layers. Wave functions and electron spectrum in a quantum limit, where the "sizes" of quasi-classical electron orbits are of the order of nano-scale distances between the layers, are determined. AC infrared measurements to investigate Fermi surfaces and to test Fermi liquid theory in Q2D organic and high- T_c materials in high magnetic fields, $H \simeq 10 - 45 T$, are suggested.

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Layered quasi-one-dimensional (Q1D) and quasi-two-dimensional (Q2D) organic conductors exhibit unique magnetic properties [1-6]. Recently [4], it has been understood that most of them can be explained in terms of effective space dimensionality crossovers for electron wave functions in a magnetic field. In their simplest forms, $3D \rightarrow 2D$ dimensional crossovers were suggested to explain field-induced spin-density wave phases [5-8,1] and to predict reentrant superconductivity (RS) phenomenon [8-10] .

In particular, in a Q2D conductor with electron spectrum,

$$\epsilon(\mathbf{p}) = \epsilon_{\parallel}(p_x, p_y) + 2t_{\perp} \cos(p_z d) , \quad t_{\perp} \ll \epsilon_{\parallel}(p_x, p_y) \sim \epsilon_F , \quad (1)$$

in a parallel magnetic field,

$$\mathbf{H} = (0, H, 0) , \quad \mathbf{A} = (-Hz, 0, 0) , \quad (2)$$

quasi-classical electron trajectories, determined by the equations of motion,

$$dp_z/dt = ev_x(p_x, p_y)H/c , \quad v_x(p_x, p_y) = d\epsilon_{\parallel}(p_x, p_y)/dp_x , \quad (3)$$

become periodic and restricted along \mathbf{z} -axis:

$$\begin{aligned} z(t, p_x, p_y, H) &= l_{\perp}(p_x, p_y, H) \cos[\omega_c(p_x, p_y, H)t] , \\ l_{\perp}(p_x, p_y, H) &= 2t_{\perp}/\omega_c(p_x, p_y, H) , \quad \omega_c(p_x, p_y, H) = ev_x(p_x, p_y)Hd/c . \end{aligned} \quad (4)$$

Periodic along \mathbf{z} -axis electron trajectories (4) correspond to "two-dimensionalization" of electron wave functions [9]. It is important that quasi-classical (QC) $3D \rightarrow 2D$ dimensional crossovers happen at weak magnetic fields [5,9], where the "sizes" of electron orbits (4) are much larger than the inter-plane distances, $l_{\perp}(p_x, p_y, H) \gg d$. For instance, these QC crossovers are responsible for novel type of cyclotron resonance (CR) on open orbits [11-15]. In particular, Kovalev et al. [15] have suggested a new method to investigate Q2D Fermi surfaces (FS) by means of CR [11-14] and studied FS in organic conductor κ -(ET)₂I₃. For theoretical justification of the method, they used QC kinetic equation, which is appropriate under experimental conditions [15], where $l_{\perp}(p_x, p_y, H) \geq d$ at $H \simeq 1 - 5 T$.

Meanwhile, in high experimental fields, $H \simeq 10 - 45 T$, typical "sizes" of electron orbits (4) become less than inter-layer distances [8],

$$l_{\perp}(p_x, p_y, H) \leq d \simeq 10 - 30 A , \quad (5)$$

in a number of Q2D organic and high- T_c materials. Under condition (5) [which we call quantum limit (QL)], theoretical methods used so far [5-15] are not justified. On the other hand, it is known that existence or not of Q2D Fermi surfaces is one of the main problems

in the area of high- T_c and organic materials [16]. In this context, it is important to suggest quantum mechanical variant of Kovalev et al. method [15] to investigate Q2D FS in high fields (i.e., in QL case (5)), where the method is less sensitive to impurities existing in doped high- T_c materials.

The main goal of our Letter is to determine electron spectrum and wave functions in a Q2D conductor (1) in a parallel magnetic field (2). We show that, in contrast to the extended Bloch waves [17,18], all wave functions are localized on conducting planes and are characterized by some quantum numbers at $H \neq 0$. Quantization law, obtained in this Letter, is qualitatively different from well-known Landau level quantization [17,18] in a perpendicular magnetic field. As a result ac infrared properties are shown to be unusual. In particular, we use our common results to extend QC method [15] to study Q2D FS to QL case (5). We hope that this allows to test the existence of FS in numerous Q2D organic and high- T_c compounds.

To determine electron wave functions in Q2D conductor (1) in parallel magnetic field (2), we make use of QC description of electron motion within conducting (\mathbf{x}, \mathbf{y}) -planes and solve fully quantum mechanical problem for electron motion between the planes. After QC Peierls substitutions for in-plane momenta, $p_x \rightarrow p_x - (\frac{e}{c})A_x$, $p_x \rightarrow -i(\frac{d}{dx})$, $p_y \rightarrow -i(\frac{d}{dy})$ [17], one can represent electron Hamiltonian in the form:

$$\hat{H} = \epsilon_{\parallel} \left(-i\frac{d}{dx} - \frac{eHz}{c}, -i\frac{d}{dy} \right) + \frac{1}{2m} \left(-i\frac{d}{dz} \right)^2 - \frac{V}{m} \sum_{n=-\infty}^{\infty} \delta(z - dn), \quad (6)$$

where the last term introduces potential energy of crystalline lattice along \mathbf{z} -axis; $V > 0$; $\delta(\dots)$ is Dirac delta-function. Note that Hamiltonian (6) is exact one for an isotropic Q2D case. As it follows from general theory [17], the above mentioned method disregards only corrections of the order of $\omega_c^2(p_x, p_y, H)/\epsilon_F$ to electron energy for arbitrary function $\epsilon_{\parallel}(p_x, p_y)$. It is seen from Eqs. (4),(5) that QL condition corresponds to $t_{\perp} \sim \omega_c(p_x, p_y, H)$, and, thus, these corrections are of the order of $t_{\perp}^2/\epsilon_F \ll \omega_c(p_x, p_y, H)$, where $\omega_c(p_x, p_y, H)$ is a characteristic energy scale in a magnetic field. Therefore, Hamiltonian (6) allows to study both QC and QL (5) dimensional crossovers.

Arbitrary solution of the Schrodinger equation for Hamiltonian (6) can be written as

$$\Psi_{\epsilon}(x, y, z) = \exp(ip_x x) \exp(ip_y y) \Psi_{\epsilon}(p_x, p_y; z), \quad (7)$$

which corresponds to free electron motion within (\mathbf{x}, \mathbf{y}) -planes. After substitution of Eq. (7) into Hamiltonian (6), it can be rewritten as follows:

$$\hat{H} = \epsilon_{\parallel} \left(p_x - \frac{eHz}{c}, p_y \right) - \left(\frac{1}{2m} \right) \frac{d^2}{dz^2} - \frac{V}{m} \sum_{n=-\infty}^{\infty} \delta(z - dn). \quad (8)$$

By expanding in-plane energy in powers of H , it is easy to make sure that Schrodinger equation for Hamiltonian (8) with the same accuracy can be expressed as:

$$\left[-\left(\frac{1}{2m}\right) \frac{d^2}{dz^2} - \omega_c(p_x, p_y, H) \frac{z}{d} - \frac{V}{m} \sum_{n=-\infty}^{\infty} \delta(z-dn) \right] \Psi_{\epsilon}(p_x, p_y; z) = [\epsilon - \epsilon_{\parallel}(p_x, p_y)] \Psi_{\epsilon}(p_x, p_y; z) . \quad (9)$$

It is possible to prove [19] that, if one uses tight binding approximation for solutions of Eq.(9),

$$\Psi_{\epsilon}(p_x, p_y; z) = \sum_{m=-\infty}^{\infty} A_m(p_x, p_y) \Phi_{\epsilon_0}(z - dm) \quad (10)$$

[where $\Phi_{\epsilon_0}(z - dm)$ is wave function of individual m -th layer at $H = 0$, corresponding to energy $\epsilon_0 < 0$, $|\epsilon_0| \sim \epsilon_F$], then one disregards only corrections of the order of $\omega_c^2(p_x, p_y, H)/[\epsilon_{\parallel}(p_x, p_y), \epsilon_0] \sim t_{\perp}^2/\epsilon_F$ to electron energy.

Therefore, equation

$$[\epsilon - \epsilon_0 - \epsilon_{\parallel}(p_x, p_y) + m\omega_c(p_x, p_y, H)]A_m(p_x, p_y) = -A_{m+1}(p_x, p_y, H)t - A_{m-1}(p_x, p_y)t , \quad (11)$$

which can be derived after substitution of wave-functions (10) into Hamiltonian (9), has the same accuracy as Hamiltonian (6) and, thus, can be used to describe $3D \rightarrow 2D$ QL dimensional crossovers (5). At given in-plane momenta p_x and p_y , Eq.(11) is equivalent to the so-called Stark-Wannier ladder equation in electric field [20]. Using Ref.[20], one can express wave functions and energy levels in the following way:

$$\begin{aligned} \Psi_N(p_x, p_y; z) &= \sum_{m=-\infty}^{\infty} J_{N-m}[2t_{\perp}/\omega_c(p_x, p_y, H)] \Phi_{\epsilon_0}(z - dm) , \\ \epsilon_N(p_x, p_y) &= \epsilon_0 + \epsilon_{\parallel}(p_x, p_y) - N\omega_c(H, p_x, p_y) , \end{aligned} \quad (12)$$

where $J_N(\dots)$ is Bessel function of N -th order [21]. [An important difference between wave functions and energy spectrum (12) and that in Ref. [20] is that the envelope functions, $J_{N-m}(\dots)$, and energy levels, $\epsilon_N(\dots)$, in Eq.(12) depend on p_x and p_y .]

Eq. (12) represents the main result of our Letter. In contrast to textbook extended Bloch waves with complex envelope, $\exp(ikz)$ [17], the envelope functions in Eq. (12) are real functions localized on the N -th conducting layer (see Fig.1). Therefore, one concludes that, in a parallel magnetic field, all wave functions are localized on layers with energy gap between two neighboring wave functions being $\omega_c(p_x, p_y, H)$. Eq.(12) is valid both in QC and QL cases.

Below, we show that quantization law (12) leads to unusual ac infrared properties and suggest a method to investigate Q2D FS. For these purposes, we calculate ac conductivity component, perpendicular to conducting layers, $\sigma_{\perp}(H, \omega)$, using known wave functions and energy spectrum (12). Let us first find matrix elements of momentum operator, $\hat{p}_z = -i\frac{d}{dz}$,

responsible for interactions between electrons and electric field, $\mathbf{E} \parallel \mathbf{z}$. It is possible to make sure that the matrix elements are non-zero only for wave functions with the same in-plane momenta, p_x and p_y , and energies $\epsilon_1 - \epsilon_2 = \pm\omega_c(p_x, p_y, H)$:

$$p_z^{N,N+1} = p_z^{N,N-1} = \int \Psi_N^*(z) \left(-i \frac{d}{dz}\right) \Psi_{N+1}(z) dz = \int \Psi_N^*(z) \left(-i \frac{d}{dz}\right) \Psi_{N-1}(z) dz = -imdt_{\perp} . \quad (13)$$

[In other words, only optical transitions between electrons with the same in-plane momenta localized on neighboring conducting layers are allowed].

To calculate $\sigma_{\perp}(H, \omega)$, we make use of the following extension [22] of Kubo formalism:

$$\sigma_{\perp}(H, \omega) = -i \frac{2e^2}{m^2 V} \sum_{N_1, N_2} \frac{|p_z^{N_1, N_2}|^2}{(E_{N_1} - E_{N_2})} \frac{[n(E_{N_2}) - n(E_{N_1})]}{(E_{N_2} - E_{N_1} - \omega - i\nu)} , \quad \nu \rightarrow 0 , \quad (14)$$

where $n(E)$ is Fermi-Dirac distribution function, V is a volume. After substituting matrix elements (13) and energy spectrum (12) in Eq.(14) and straightforward calculations, one obtains:

$$\sigma_{\perp}(H, \omega) \sim -i \int \frac{dp}{|\mathbf{v}_F(p_x, p_y)|} \left[\frac{1}{\omega_c(p_x, p_y, H) - \omega - i\nu} + \frac{1}{-\omega_c(p_x, p_y, H) - \omega - i\nu} \right] , \quad \nu \rightarrow 0 . \quad (15)$$

[Integration in Eq. (15) is made along 2D contour $\epsilon_{\parallel}(p_x, p_y) = \epsilon_F$; $\mathbf{v}_F(p_x, p_y) = d\epsilon_{\parallel}(p_x, p_y)/d\mathbf{p}$; we use the approximation $n(E_{N_2}) - n(E_{N_1}) = (E_{N_2} - E_{N_1})dn(E)/dE$ since $|E_{N_2} - E_{N_1}| = \omega_c(p_x, p_y, H) \ll \epsilon_F$].

It is convenient to write explicitly real and imaginary parts of conductivity (15):

$$\Re \sigma_{\perp}(H, \omega) \sim \int \frac{dp}{|\mathbf{v}_F(p_x, p_y)|} \left(\delta[\omega_c(p_x, p_y, H) - \omega] + \delta[\omega_c(p_x, p_y, H) + \omega] \right) = \begin{cases} \neq 0 , & \omega < \omega_c^{max}(H) \\ 0 , & \omega > \omega_c^{max}(H) \end{cases} , \quad (16)$$

$$\Im \sigma_{\perp}(H, \omega) \sim \int \frac{dp}{|\mathbf{v}_F(p_x, p_y)|} \left[\frac{1}{\omega_c(p_x, p_y, H) + \omega} - \frac{1}{\omega_c(p_x, p_y, H) - \omega} \right] , \quad (17)$$

where $\omega_c^{max}(H)$ is the maximum value of energy gap, $\omega_c(p_x, p_y, H)$, on the contour of integration (see Fig.2); integral in Eq.(17) is determined as its principle value.

The main difference between Eqs.(16),(17) and the results of Ref.[17] is that Eqs.(16),(17) are valid both in QC and QL (5) cases, whereas the results [17] are essentially QC. Another difference is that Eqs.(16),(17) describe "optical" conductivity (i.e., conductivity in the absence of impurities), in contrast to kinetic equation result [17]. From Eqs.(16),(17), it follows that ac properties in a parallel magnetic field are unusual. Indeed, integration of δ -function in Eq.(16) results in non-zero value of real part of conductivity for ac frequencies $0 < \omega < \omega_c^{max}(H)$ (see Fig.2). Therefore, electrons absorb electromagnetic waves at $0 < \omega < \omega_c^{max}(H)$ (in the absence of impurities!), in contrast to text book properties of metals [18].

Let us demonstrate that real part of conductivity (16) diverges at resonant frequency,

$$\omega = \omega_c^{max}(H) = ev_x^{max}(p_x, p_y)Hd/c . \quad (18)$$

Indeed, in the vicinity of its maximum $\omega_c(p_x, p_y, H) \simeq \omega_c^{max}(H) - A(H)|\mathbf{p}|^2$ with \mathbf{p} being momentum component perpendicular to $\mathbf{v}_F(p_x, p_y)$ at point, where $|v_x(p_x, p_y)|$ takes its maximum (see Fig.2). In this case, integral (16) can be estimated as

$$\Re\sigma_{\perp}(H, \omega) \sim \frac{1}{\sqrt{\omega_c^{max} - \omega}} , \quad \omega_c^{max}(H) - \omega \ll \omega_c^{max}(H) . \quad (19)$$

Therefore, by measuring $\omega_c^{max}(H)$ at different directions of the field one can determine angular dependence of $v_x^{max}(p_x, p_y, H)$ (see Ref.[15] and Fig.(2)). We stress, however, that the physical meaning of resonant frequency (18) at high magnetic fields (5), where electrons are almost completely localized on conducting layers (see Figs.1,2), is completely different from its kinetic equation interpretation [15,23].

To summarize, wave functions and electron spectrum of a Q2D conductor in a parallel magnetic are determined. A method to test FL picture in Q2D organic and high- T_c materials is suggested. We hope that this method is a useful experimental tool to study Fermi-liquid versus non Fermi-liquid behavior in low-dimensional compounds, especially as there have been claimed inconsistencies [24] between angular resolved photo-emission methods [16] and magneto-optical measurements in a perpendicular magnetic field [24]. We also think that $3D \rightarrow 2D$ QL dimensional crossovers and quantization law (12), suggested in the Letter, will be useful for studies of RS superconductivity [8-10] and for explanations of still unexplained phenomena observed in high parallel magnetic fields [25,26].

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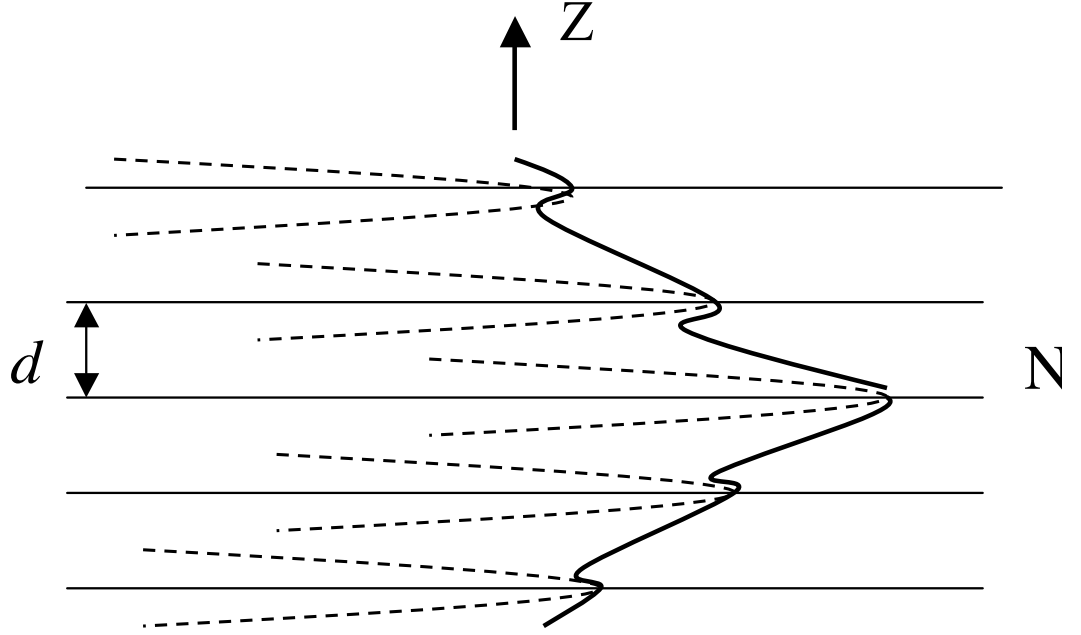


FIG. 1: Wave function of a layered Q2D conductor (1) (at fixed values of p_x and p_y), localized on N -th conducting layer in a parallel magnetic field (2), is sketched. Solid line: envelope function, $|J_{N-m}[2t_{\perp}/\omega_c(p_x, p_y, H)]|$. Dashed lines: wave functions of individual layers, $\Phi_{\epsilon_0}(z - dm)$ (see Eqs.(10),(12) and the text).

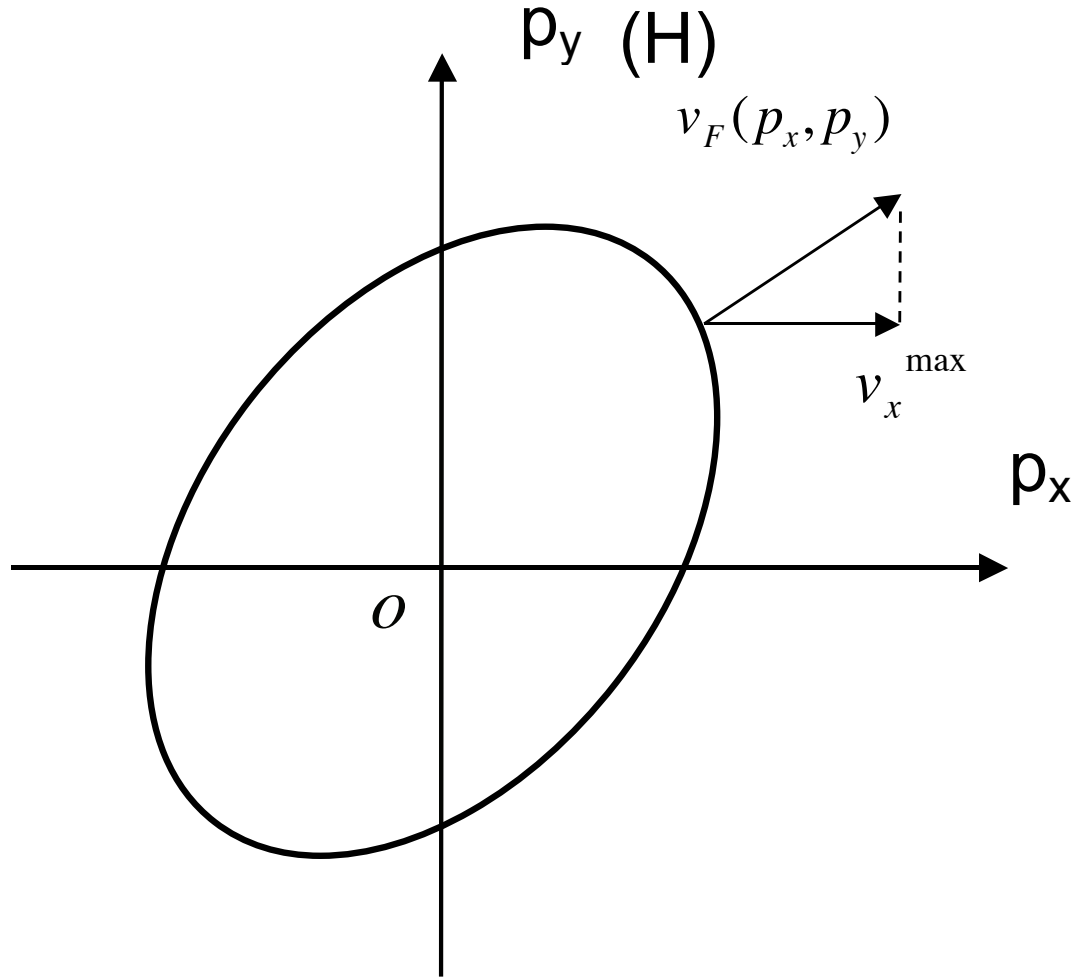


FIG. 2: Resonant frequency $\omega_c^{\max}(H)$ corresponds to the maximum value of $|v_x(p_x, p_y)|$, v_x^{\max} , on 2D Fermi surface, $\epsilon_{\parallel}(p_x, p_y) = \epsilon_F$, as it follows from Eq.(4).